Experimental report

15/02/2021

Proposal: 4-05-730 **Council:** 10/2018

Title: Spin excitations in new quantum kagome compounds of the atacamite family

Research area: Physics

This proposal is a new proposal

Main proposer: Edwin KERMARREC

Experimental team: Bruce D. GAULIN
Edwin KERMARREC
Ramender Kumar SHARMA

Local contacts: Jean Marc ZANOTTI

Samples: MgCu3(OD)6(NO3)2
ZnCu3(OD)6(NO3)2
NiCu3(OD)6(NO3)2

Instrument	Requested days	Allocated days	From	To
IN6-SHARP	6	4	19/07/2019	23/07/2019

Abstract:

Geometrically frustrated magnets provide a plethora of opportunities to realize exotic ground states. Of these, spin-liquid state has spurred the attention of researchers because of its interesting properties and markedly different elementary excitations from that of conventional magnets. The kagome lattice, made of corner sharing triangles, was identified as the best candidate among 2D models, and there is no surprise that many efforts have been pursued to understand the spin dynamics of the S=1/2 Heisenberg antiferromagnetic kagome model.
We have recently succeeded in synthesizing new kagome members belonging to the atacamite family, MCu3(OH)6(NO3)2 where M = Zn, Mg or Ni, of the "kapellasite-type" structure, that are realizations of the related J1-J2-Jd Heisenberg model. They offer the opportunity to confront experimental results to the theoretical phase diagram of such model established recently, where exotic non coplanar states sis next to spin liquid and valence bond crystal phases.

Experimental report 04-05-730

Spin excitations in new quantum kagome compounds of the atacamite family

S. Bhattacharya[1], R. Kumar[1], J.-M. Zanotti[2], B. D. Gaulin[3], P. Puphal[4] and E. Kermarrec[1]

[1] *Université Paris-Saclay, CNRS, Laboratoire de Physique des Solides, 91405, Orsay, France*
[2] *Laboratoire Léon Brillouin, CEA, CNRS, Université Paris-Saclay, F-91191 Gif-sur-Yvette, France*
[3] *Departement of Physics and Astronomy, McMaster University, Hamilton (ON, Canada)*
[4] *Paul Scherrer Institut, Villigen, Switzerland*

In search for spin-liquids, a number of frustrated lattices have been examined since the 90s and the kagome lattice, made of corner sharing triangles, was identified as the best candidate among 2D models. Research efforts have led to the synthesis of a variety of $S = 1/2$ kagome compounds, derived mostly from the atacamite mineral family (Cu^{2+}, $S = 1/2$). We have recently succeeded in synthesizing new kagome members belonging to this atacamite family, $M\mathrm{Cu_3(OD)_6(NO_3)_2}$ where M = Zn, Mg or Ni. As shown by previous experiments on kapellasite and haydeite, inelastic neutron scattering is crucial to determine the ground state and the hamiltonian parameters of these new kagome magnets.

Experimental details

We performed inelastic neutron scattering on the IN6-SHARP spectrometer on three deuterated powder samples of $M\mathrm{Cu_3(OD)_6(NO_3)_2}$ (where M = Zn, Mg or Ni) and on an empty can for background purposes. Each powder sample was wrapped inside aluminium foil and loaded into an aluminium cylinder of 1.5 cm diameter with a height of 5 cm and mounted inside an Orange cryostat. The chosen incident wavelength was $\lambda = 5.12$ Å. During the experiment we could measured $S(q, \omega)$ intensity maps for various temperatures between 30 K and 1.5 K in zero field.

Main results

$\mathbf{ZnCu_3(OD)_6(NO_3)_2}$. Fig.1 shows the temperature evolution of the dynamical structure factor $S(q, \omega)$ for selected temperature above and below the transition temperature $T_N \simeq 8$ K. Note that the narrow linear inelastic feature below 1 meV extending up to 1.2-1 is due to spurious elastic reflexions. It displays a broad quasi-elastic scattering signal above T_N located closeto the M point of the Brillouin zone, a very peculiar position that points to non-coplanar correlations of the cuboc2 chiral phase, similarly to the Zn-sister kapellasite compound. However, here for $T < T_N$ the low-Q intensity becomes inelastic in agreement with the formation of (powder-averaged) spin-waves. Concomitantly, elastic magnetic Bragg peaks appears within our poor Q-resolution. This is a very exciting result as it would be the first evidence of such exotic non-coplanar cuboc2 ordered phase, predicted from symmetry argument only, if confirmed.

$\mathbf{NiCu_3(OD)_6(NO_3)_2}$. Fig.2 shows the temperature evolution of the dynamical structure factor $S(q, \omega)$ for selected temperature above and below $T* \simeq 8$ K, the temperature of the broad maximum observed in magnetic susceptibility measurement. However, for this Ni-variant no clear gapped spectrum is observed upon cooling. We rather observe a broad, quasi-elastic signal peaked around ~ 0.6 Å that persists well below T^* down to 1.5 K. In the elastic channel, a non-resolution limited peak appears below 5 K centered at ~ 0.5 Å, that is perhaps associated with the formation of a short-range

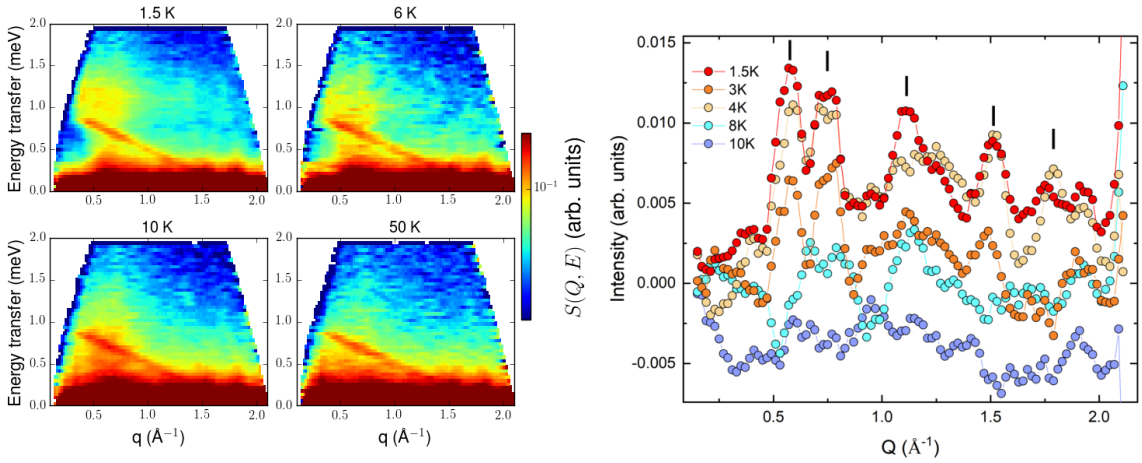

Figure 1: (Left panel) Temperature evolution of $S(q,\omega)$ for $ZnCu_3(OD)_6(NO_3)_2$ above and below $T_N \simeq$ 8 K. (Right panel) Temperature dependence of the elastic integrated-energy cut (E=[-0.1,0.1]meV). A 20K-dataset has been subtracted off.

magnetic order.

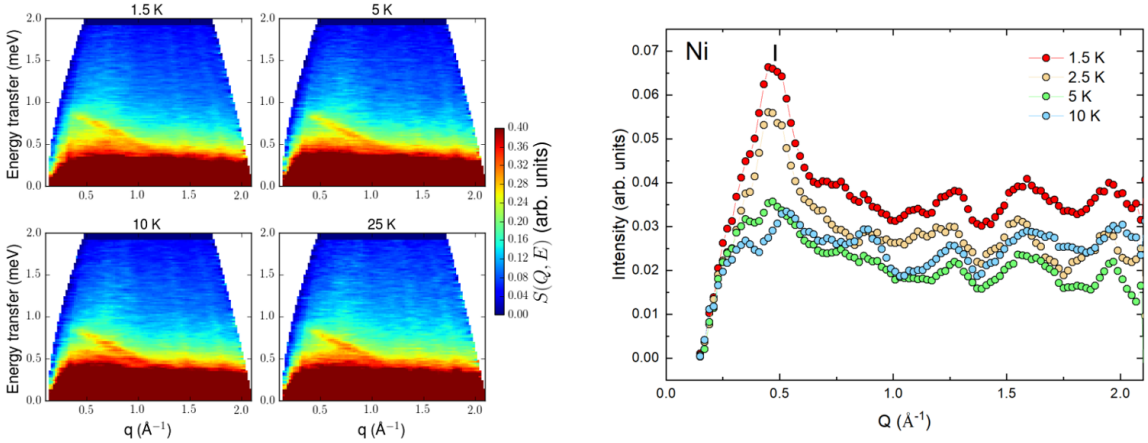

Figure 2: (Left panel) Temperature evolution of $S(q,\omega)$ for $NiCu_3(OD)_6(NO_3)_2$. (Right panel) Temperature dependence of the elastic integrated-energy cut (E=[-0.1,0.1]meV). A 25K-dataset has been subtracted off.

$MgCu_3(OD)_6(NO_3)_2$. Fig.3 shows the temperature evolution of the dynamical structure factor $S(q,\omega)$ for the Mg-variant, for the only two temperatures measured in the remaining time of the experiment. In contrast to the two other compositions, the inelastic spectrum is mainly composed of a flat, quasi-elastic signal extending up to $\sim$ 1 meV and no clear sign of magnetic elastic contribution is observed in the comparison between the 1.5 K and the 30 K datasets.

In conclusion, the three probed compositions show very different elastic and inelastic contributions which could reflect different magnetic phases selected by different scheme of magnetic interaction. Complementary experiments and theoretical analysis, such as high temperature series expansions, are planned to further help to determine the values of the exchanges for each compound and confirms this scenario.

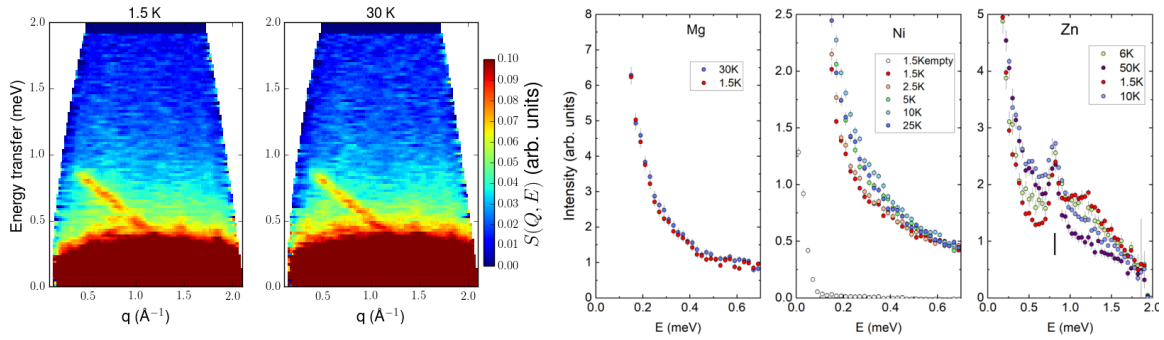

Figure 3: (Left panel) Temperature evolution of $S(q,\omega)$ for $MgCu_3(OD)_6(NO_3)_2$. (Right panel) Temperature dependence of the Q-integrated cut ($Q = 0.5 \pm 0.05$ Å^{-1}) for all compositions.